# Low temperature electronic properties of $Sr_2RuO_4$ III: Magnetic fields

Ralph Werner
*Physics Department, Brookhaven National Laboratory, Upton, NY 11973-5000, USA and*
*Institut für Theorie der Kondensierten Materie, Universität Karlsruhe, 76128 Karlsruhe, Germany*
(February 1, 2008)



Based on the microscopic model introduced previously the observed specific heat and ac-susceptibility data in the superconducting phase in $Sr_2RuO_4$ with applied magnetic fields are described consistently within a phenomenological approach. Discussed in detail are the temperature dependence of the upper critical fields $H_{c2}$ and $H_2$, the dependence of the upper critical fields on the field direction, the linear specific heat below the superconducting phase transition as a function of field or temperature, the anisotropy of the two spatial components of the order parameter, and the fluctuation field $H_{\rm p}$.

## I. INTRODUCTION

The discovery of superconductivity below $T_c \sim 1$ K in $Sr_2RuO_4$ has quickly triggered a large amount of interest[1] because of the unconventional properties[2] and the initially proposed analogy[3] to $^3$He. The material is tetragonal at all temperatures.[4] The three bands cutting the Fermi level with quasi two-dimensional Fermi surfaces[5,6] can be mainly associated with the three $t_{2g}$ orbitals of the $Ru^{4+}$ ions.[7,2] The transport is Fermi liquid like[8–10] for $T_c < T < 30$ K and strongly anisotropic along the $c$ axis.[1] The enhanced specific heat, magnetic susceptibility and electronic mass indicate the presence of significant correlations.[1,6,11] The specific heat,[12–14] thermal conductivity,[15] and nuclear quadrupole resonance (NQR)[10] are consistent with two-dimensional gapless fluctuations in the superconducting phase. For a more detailed overview see Refs. 2,16.

The present work is the last part of a series of three. In part I (Ref. 16) the quasi one-dimensionality of the kinetic energy of the $d_{zx}$ and $d_{yz}$ electrons has been used to derive an effective microscopic model. At intermediate coupling the interaction leads to a quasi one-dimensional model for the magnetic degrees of freedom and two-dimensional correlations in the charge sector. The normal phase properties are described consistently. In part II (Ref. 17) it is shown that the pair-correlations are enhanced by inter-plane umklapp-scattering as a consequence of the body centered crystal structure. The inter-plane coupling can be treated mean-field like.[18] The order parameter is a spin triplet with two slightly anisotropic spatial (flavor) components. The model consistently can account for the experimental data concerning the temperature dependence of the pair density, specific heat, NQR and muon spin relaxation ($\mu$SR) times and susceptibility. The crucial difference to previous approaches[3,19–21] is that the relevant internal degrees of freedom of the superconducting order parameter have been extracted from the material specific microscopic model.

The present paper is based on a phenomenological model that is consistent with the results derived in Refs. 16 and 17. The upper critical field[13,22–24] can be described by mean-field theory. The specific heat data[13] are in quantitative agreement with the presence of a cubic term in the Landau expansion of the free energy (Sec. II). The various observed critical fields as well as their dependence on the direction of the applied field[22–24] are well described within the framework of the flavor degrees of freedom of the model (Sec. III).

## II. MEAN-FIELD THEORY

In Ref. 17 it has been shown that the thermodynamic properties of $Sr_2RuO_4$ near the superconducting phase transition are well described by a Landau expansion in the pair excitation energy gap $\Delta$.

$$F_\Delta = F_0 - A\ t\ \Delta^2 + D\ \Delta^3 + B\ \Delta^4 + \mathrm{O}(\Delta^5) \qquad (1)$$

Here $t = 1 - T/T_c \ll 1$ is the reduced temperature. The presence of the term $\sim \Delta^3$ can be motivated by effectively integrating out the two-dimensional, gapless order parameter fluctuations.[17] Gauge invariance is preserved since $\Delta \propto |e^{i\phi_{\rm G}}\langle P\rangle|$ for pair operators $P$ and arbitrary gauge fields $\phi_{\rm G}$. To be consistent with the notation of Ref. 17 the coefficients are given as $A = N\,(64\ V_0)^{-1}$, $B = N\frac{b}{(8\ V_0)^4}$, $D = N\frac{s_0^2}{128\ \overline{v}_{\rm eff}^2}$. The parameters define the number of Ru ions $N$, the effective velocity of the elementary electronic excitations above the gap $\overline{v}_{\rm eff} \approx 22$ K, the numerical prefactor $s_0 \approx 4.7$ K, and the interplane pair hopping potential $V_0 \approx 6$ K. If one assumes that the excitations of the magnetic degrees of freedom of the order paramter are gapped through spin-flavor coupling the numbers are $\overline{v}_{\rm eff} \approx 38$ K and $s_0 \approx 2.7$.

Comparing with experiments[25,26] the superconducting gap has been found to be linear in the reduced temperature[17] over a rather large interval, i.e.,

$$\Delta(T)\big|_{0\leq t<0.5} \approx \frac{2A}{3D}\ t \approx 0.8\ V_0\ t\,. \qquad (2)$$

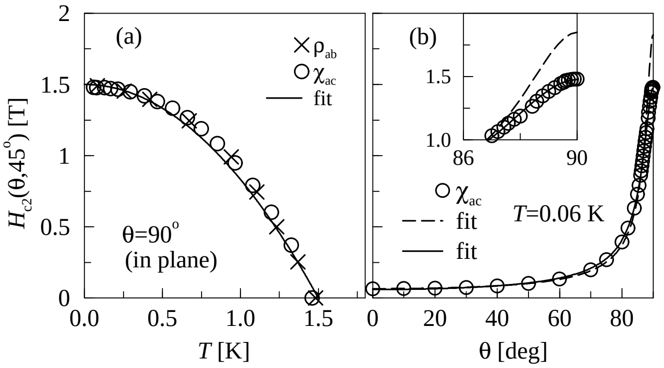


FIG. 1. *Upper critical magnetic fields. Circles are from ac-susceptibility measurements Ref. 24, crosses are from resistivity measurements Ref. 28. (a) $H_{\rm cm}$ along [110] as a function of temperature, fit from Eq. (3). (b) $H_{\rm c2}(\theta, 45°)$ as a function of the polar angle $\theta$. The dashed line is a fit from Eq. (4), the solid line is a fit from Eq. (5). The inset is an enlargement of the region $\theta \sim 90°$.*

Since $\Delta \geq 0$ the phase transition described by Eqs. (1) and (2) is of third order in the sense of Ehrenfest's definition.[27]

### A. Upper critical fields $H_{\rm c2}$

The field above which superconductivity disappears is denoted $H_{\rm c2}(\theta, \phi)$. The azimuthal $\phi$ and polar $\theta$ angles are defined as $H(0, \phi) \| [001]$ and $H(90°, 0) \| [100]$. The critical field is maximal along [110],[23,24] i.e., $H_{\rm cm}(T) = H_{\rm c2}(90°, 45°)$. $H_{\rm cm}(T)$ is plotted in Fig. 1(a) as derived from ac-susceptibility measurements (circles)[24] and resistivity data (crosses).[28] The fit to the phenomenological curve[29]

$$\frac{H_{\rm cm}(T)}{H_{\rm cm}(T=0)} = 1 - \left(\frac{T}{T_c(H=0)}\right)^2 \qquad (3)$$

with $H_{\rm cm}(T = 0) = 1.5$ T and $T_c(H = 0) = 1.5$ K reproduces the experimental data satisfactorily [full line in Fig. 1(a)]. The deviation of Eq. (3) from the BCS predictions $\lim_{T\to 0} \frac{H_{\rm cm}(T)}{H_{\rm cm}(T=0)} \approx 1 - 1.06 \frac{T^2}{T_c^2(H=0)}$ and $\lim_{T\to T_c} \frac{H_{\rm cm}(T)}{H_{\rm cm}(T=0)} \approx 1.74[1 - \frac{T}{T_c(H=0)}]$ is smaller than the experimental uncertainty.

The experimental uncertainties in the determination of $H_{\rm cm}$ lie in part in the ambiguities between $T_{\rm c}$ and the "mid transition" value $T_{\rm cm}$ due to the linear order parameter as discussed in Ref. 17. In Ref. 24 the possible violation of the Werthamer-Helfand-Hohenberg (WHH) formula[30] is discussed. The WHH formula relates $H_{\rm cm}(T = 0)$ and the derivative $dH_{\rm cm}(T)/dT|_{T=T_c}$. Since the experimental estimate relies solely on the two points closest to $T_c(H = 0)$ the discrepancies must very likely be attributed to experimental uncertainties.

Figure 1(b) shows the critical field $H_{\rm c2}(\theta, 45°)$ as a function of the polar angle. The fit from the Landau-Ginzburg anisotropic effective mass approximation[31] defined by

$$H_{\rm c2}(\theta, 45°) = \frac{H_{\rm c2}(\theta = 0, 45°)}{|\cos\theta|\sqrt{1 + \tan^2\theta / R_m^2}} \qquad (4)$$

gives an effective mass ratio of $R_m \approx 28$ [dashed line in Fig. 1(b)].

While the experimental data[24] are well reproduced for $\theta < 87°$, the fit function overestimates the in-plane critical field at $\theta = 90°$ by roughly 20% [inset of Fig. 1(b)]. Such an enhancement of the in-plane coupling to the magnetic field is expected from the 20% easy plane anisotropy of the spin-one Cooper pairs that was implied from the $\mu$SR relaxation time anisotropy.[17] The easy plane configuration can be incorporated into Eq. (4) by enhancing the in-plane coupling by a factor $g_\| = 1.23$.

$$H_{\rm c2}(\theta, 45°) = \left| \begin{pmatrix} g_\| \sin\theta \\ \cos\theta \end{pmatrix} \right| \frac{H_{\rm c2}(\theta = 0, 45°)}{|\cos\theta|\sqrt{1 + \tan^2\theta / R_m^2}} \qquad (5)$$

The experimental data are then well described with $R_m = 20$ [solid line in Fig. 1(b)]. As expected this value excellently matches the in-plane to out-of-plane hopping ratio $R_m = t_0/t_\perp$.[16]

Both the temperature dependence of $H_{\rm cm}$ and the angular dependence of $H_{\rm c2}(\theta, 45°)$ strongly support the applicability of the mean-field approach.

### B. Landau theory with magnetic field

Using Eqs. (1) and (2) the specific heat just below the superconducting transition becomes

$$\frac{C_s|_{t\ll 1}}{T_{\rm c}} = \frac{C_n}{T_{\rm c}} + \frac{8}{9}\frac{A^3}{T_{\rm c}^3 D^2}\,(T_{\rm c} - T) - {\rm O}(t^2)\,. \qquad (6)$$

The critical temperature depends on the magnetic field $T_{\rm c} = T_{\rm c}(H)$. The normal phase contribution is constant with $\frac{C_n}{T_{\rm c}} \approx 37.5\,\frac{\rm mJ}{\rm K^2 mol}$. The coefficient $\frac{8}{9}\frac{A^3}{T_{\rm c}^3 D^2}$ can be determined from the measurements in Ref. 14 as a function of the applied magnetic field. The results are plotted in Fig. 2 (symbols) and can be fitted by

$$\gamma = \frac{8}{9}\frac{A^3}{T_{\rm c}^3 D^2} = 310\sqrt{1 - \frac{H}{H_{\rm cm}(0)}}\;\frac{\rm mJ}{\rm K^3 mol} \qquad (7)$$

with $H_{\rm cm}(0) = 1.5$ T (full line).

The reduced temperature at a given temperature is altered as the magnetic field is changed, i.e., $t = t(H)$, because of the resulting change in the critical temperature. The latter can be obtained by inverting Eq. (3) to give $T_{\rm c}(H)$. Defining the reduced field $h(T) = 1 - \frac{H}{H_{\rm cm}(T)}$

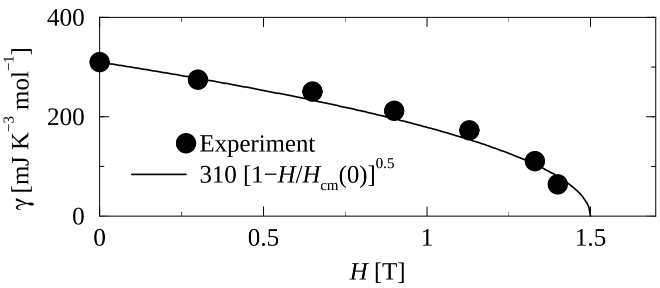


FIG. 2. *Coefficient $\gamma = \frac{8}{9}\frac{A^3}{T_c^3 D^2}$ as determined from the measurements in Ref. 14 as a function of the applied magnetic field (symbols). The full line [Eq. (7)] is the fit consistent with the mean-field approach.*

at a given temperature and expanding Eq. (3) for $t \ll 1$ one finds the relation

$$t(H)\ \frac{T_c^2(H)}{T_c^2(0)} \approx \frac{1}{2}\frac{H_{\rm cm}(T)}{H_{\rm cm}(0)}\ h(T) \tag{8}$$

between the reduced temperature and the reduced field. Using Eq. (8) and the fit of the coefficient $\gamma$ as given in Eq. (7) the specific heat as a function of the magnetic field becomes at a given temperature for $0 \le h \ll 1$

$$\frac{C_s|_{h\ll1}}{T} = \frac{C_n}{T} + \frac{155\ \mathrm{mJ}}{\mathrm{TK^2mol}}\ (H_{\rm cm} - H) - \mathrm{O}(h^2)\,. \tag{9}$$

From the experimental results in Ref. 14 one finds for $0.4\mathrm{K} < T < 1.5\mathrm{K}$ values for the linear field coefficient of 140 to 160 $\frac{\mathrm{mJ}}{\mathrm{TK^2mol}}$ in excellent agreement with the prediction.

For $H > 1.4$ T or $T_c < 0.4$ K corrections to the fit of $\gamma$ must be expected. The data in Fig. 2 are consistent with a faster drop of $\gamma$ for $H > 1.4$ T than anticipated by the fit from Eq. (7). At the the same time the region of validity of Eq. (8) is reduced to extremely small values of $h$. For $T \to 0$ the term linear in $H$ in Eq. (9) then becomes negligible. The higher order terms in Eq. (9) are negative and are consistent with the sharp drop of the specific heat with $h$ for $T \to 0$.[14] Detailed specific heat measurements in the range 1.4 T $< H <$ 1.5 T are desirable for further clarification.

Using the definition Eq. (7) and Eq. (3) to express $T_c(H)$ as a function of the magnetic field gives

$$D(H) = 4.88\ \mathrm{K}\ \sqrt{\frac{A}{T_c(0)}}^{\,3}\ \left(1 - \frac{H}{H_{\rm cm}(0)}\right)^{-1}. \tag{10}$$

Assuming that the coefficient $A$ is independent[17] of the field $H$ and replacing $D \to D(H)$ in Eq. (2) one finds

$$\Delta_H(T)\big|_{t\ll1} \approx \Delta_0\ \sqrt{1 - \frac{H}{H_{\rm cm}(0)}}\ \frac{T_c(H) - T}{T_c(0)}\,, \tag{11}$$

with $\Delta_0 = 4.9$ K. Applying Eq. (8) yields

$$\Delta_T(H)\big|_{h\ll1} \approx \Delta_0\ \frac{H_{\rm cm}(T) - H}{H_{\rm cm}(0)}\,. \tag{12}$$

Equations (10) – (12) only are meaningful under the assumption that the coefficient $A$ is independent of the field $H$. They yield meaningful results consistent with experiments within the present approach justifying the assumption made. An unambiguous quantitative test of Eqs. (11) and (12) can be obtained via excess current measurements.[17,26]

### C. Low temperature specific heat

For $T \to 0$ and $H < H_{\rm sg} \approx 0.12$ T the specific heat of $Sr_2RuO_4$ shows a sharp linear increase as a function of the applied magnetic field.[14] This phenomenon can be easily understood in terms of a small gap in the magnetic excitation spectrum of internal magnetic degrees of freedom of the superconducting order parameter which is induced by spin-flavor coupling.[17,32] The applied magnetic field splits the three components of the spin triplet described by the SO(3) vector[17] $\overline{\mathbf{\Omega}}_{\rm s}$ and the lowest spin state is decreased in energy until the spin gap is closed. The spin gap can be estimated as $\Omega_A \approx \mu_{\rm B} H_{\rm sg} = 0.08$ K. This value is consistent with an estimate obtained via the analogy[33] to $^3$He-$A$ and suggests that for temperatures $T > 0.1$ K the magnetic excitations of the Cooper pair moments can be considered as gapless.

For $H > 0.12$ T and at low temperatures $0.1\ \mathrm{K} \le T \ll T_c$ the experimental specific heat can be described by a contribution linear in $T$ and a contribution quadratic in $T$.[14] The quadratic contribution stems from the quasi two-dimensional gapless fluctuations of the internal degrees of freedom of the order parameter. In the presence of a magnetic field the magnetic degeneracy of the magnetic pair order parameter components is lifted by the Zeeman splitting. The non-linear sigma model describing the gapless fluctuations of the order parameter components is thus reduced from SO(3)⊗SO(2) to one channel or SO(2) symmetry. The quadratic specific heat contribution then is [c.f. Eq. (70) in Ref. 17]

$$C_s^{(2)}|_{T\to0} = \frac{3\,\zeta(2)}{\pi}\ \frac{T^2}{(v_\nu)^2} \approx 1.15\ \frac{T^2}{(\overline{v}_{\rm eff})^2}\,. \tag{13}$$

Equation (13) predicts a specific heat contribution quadratic in temperature that is basically independent of the magnetic field. This result is consistent with experiments (c.f. Fig. 6 in Ref. 14) where the coefficient has been determined for 1.4 T $\ge H >$ 0.12 T as $C^{(2)}_{s,\rm exp}|_{T\to0} \approx 44\ \frac{\mathrm{mJ}}{\mathrm{K^3mol}}\ T^2$. Comparison with Eq. (13) leads to $\overline{v}_{\rm eff} \approx 15$ K which compares reasonably well with the value of 22 K determined in the absence of a magnetic field.[17] The change of the excitation velocity is conceivable in a magnetic field as well as quadratic temperature contributions from the vortex lattice.[34,35]

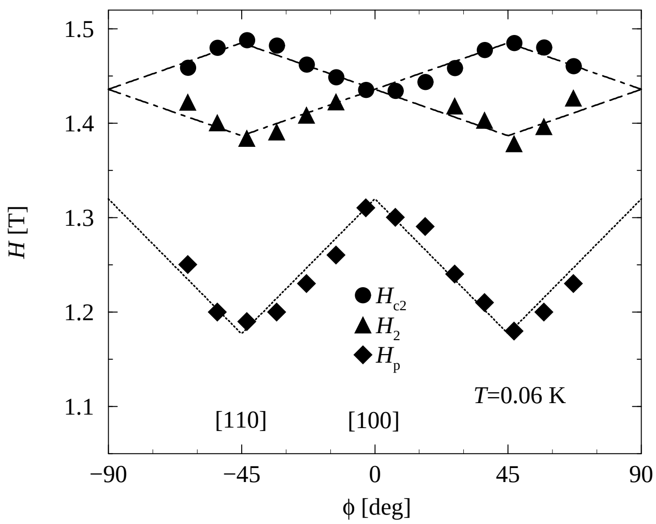


FIG. 3. *In-plane critical magnetic fields. Symbols are from ac-susceptibility measurements Ref. 24. The broken line is $H_{c,x}(\phi)$, the dash-dotted line $H_{c,y}(\phi)$ as defined in Eq. (15) with $H_{c,u} = 1.44$ T and $H_{c,a} = 0.05$ T. The dotted line is the crossover field $H_p$ as defined in Eq. (22) that marks the onset of flavor fluctuations.*

The linear contribution to the specific heat for $H > 0.12$ T stems form the excitations of the (non-degenerate) lowest spin-triplet state discussed above as well as from the vortex lattice.

## III. CRITICAL MAGNETIC FIELDS

Experimentally different critical magnetic fields have been observed in $Sr_2RuO_4$.[1,14,22–24,36,37] So far the physical implications have been discussed controversially.

### A. In-plane anisotropy of $H_{c2}$

The ac-susceptibility measurements reveal an angular alternation of the in-plane upper critical field $H_{c2}(\phi)$ with four fold symmetry[23,24] (full circles in Fig. 3). A second transition is observed at $H_2(\phi) \le H_{c2}(\phi)$ also with four fold symmetry but out-of-phase modulation (full triangles in Fig. 3). The polar angle is set to $\theta = 90°$ throughout this subsection.

In Ref. 17 it has been shown that the superconducting order parameter in $Sr_2RuO_4$ has two spatially slightly anisotropic components described by the SO(2) vector $\mathbf{\Omega}_f$. The magnetic field couples strongest to the order parameter component with dominant superconducting correlations perpendicular to the field.

Generalizing Eq. (12) to a two component vector $\mathbf{\Delta}_T \sim \mathbf{\Omega}_f$ the components $\nu = x, y$ of the order parameter become

$$\Delta_{T,\nu}(H,\phi) = \frac{\Delta_0}{\sqrt{2}}\,\frac{H_{c,\nu}(\phi,T) - H(\phi)}{H_{c,\nu}(\phi,0)}\,. \qquad (14)$$

Note that the superconducting correlations of the component $\Delta_{T,x}$ are largest along [110] while for $\Delta_{T,y}$ they are largest along $[\bar{1}10]$.[17] The critical fields can be parameterized into a homogeneous part and an alternating part[20] reflecting the two-fold symmetry of the order parameter components.

$$H_{c,{}^x_y}(\phi,T) = H_{c,u}(T) + H_{c,a}(T)\ f(\pm\sin 2\phi) \qquad (15)$$

The two components $H_{c,x}$ and $H_{c,y}$ only differ by a phase shift $\phi \to \phi + \pi/2$ reflecting the symmetry relation of $\Delta_{T,x}$ and $\Delta_{T,y}$. The total order parameter $|\mathbf{\Delta}_T(H \to 0)|$ has the four-fold symmetry of the underlying tetragonal lattice.[17]

An appropriate choice for the anisotropy function is

$$f_\alpha(\sin 2\phi) = \frac{\arcsin(\alpha \sin 2\phi)}{\arcsin \alpha} \qquad (16)$$

that interpolates between a sinusoidal function for $\alpha \to 0$ and a zig-zag function for $\alpha \to 1$. The fits in Fig. 3 for $H_{c,x}$ (dash-dotted line) and for $H_{c,y}$ (dashed line) reveal that the experimental data are best described by $\alpha \approx 1$. The parameters are determined to be $H_{c,0}(T \to 0) = 1.44$ T and $H_{c,a}(T \to 0) = 0.05$ T. They are consistent with the observations made by thermal conductivity measurements.[36,38] Note that the definition of "$H_2$" in Ref. 36 differs from the one made here.

The ratio $2H_{c,a}/H_{c,u} \approx 0.07$ is consistent with the observed anisotropies in the thermal conductivity in the superconducting phase.[39] A more detailed and quantitative comparison with the thermal conductivity data requires a more involved analysis of the transport properties of the model discussed here. It must include the symmetry breaking effect of the temperature gradient.[17]

The fields $H_{c,x}$ and $H_{c,y}$ can be combined to give the initial interpretation of the transitions in terms of

$$H_{c2}(\phi) = \sup\left[H_{c,x}(\phi), H_{c,y}(\phi)\right] \qquad (17)$$

and

$$H_2(\phi) = \inf\left[H_{c,x}(\phi), H_{c,y}(\phi)\right]\,, \qquad (18)$$

which then reflect the four-fold symmetry of the lattice.

The angular variation of the order parameter components in Eq. (14) with $\alpha = 1$ is non-analytic at $\phi_n = \frac{\pi}{4}(2n+1)$. This is consistent with the presence of quasi one-dimensional correlations along the diagonals of the tetragonal reciprocal lattice as predicted by the microscopic model for $Sr_2RuO_4$.[16] A more detailed analysis of how the quasi one-dimensional correlations determine the order parameter would require the study of the two-dimensional sine-Gordon actions[17] that determine the full Eliashberg[40] equation. This is not evident and must be left for future studies. Experimentally it would be interesting to study the in-plane anisotropy as shown in Fig. 3 at various temperatures in order to obtain the temperature dependence of $\alpha$.

### B. In-plane anisotropy of $H_{\mathrm{p}}$

The field $H_{\mathrm{p}}$ is defined by the upper edge of a shoulder formed by an increase of the ac-susceptibility with increasing reduced field $h$ as measured in Ref. 24. The experimental data are shown by the full diamonds in Fig. 3 and suggest a close relation of $H_{\mathrm{p}}$ to the critical order parameter fields $H_{\mathrm{c},x}$ and $H_{\mathrm{c},y}$.

With an alternating field of 0.05 mT at frequencies of 700-1000 Hz the ac-susceptibility measurements probe very low energy magnetic excitations of the system.[24] Within the framework of the underlying microscopic model the magnetic subsystem[16] has an energy scale of $\overline{v}_{\mathrm{eff}} \sim 22$ K. Since typical magnetic fields $H \sim 1$ T $\sim 0.7$ K $\ll \overline{v}_{\mathrm{eff}}$ its contribution remains essentially unaffected.[41] The spin triplet components $\overline{\mathbf{\Omega}}_{\mathrm{s}}$ of the order parameter is Zeeman split in a magnetic field (see also Sec. II C). On the other hand, interaction terms in the microscopic model couple the magnetic degrees of freedom to the flavor degrees of freedom $\mathbf{\Omega}_{\mathrm{f}}$ of the order parameter.[17] The ac-susceptibility is thus an indirect probe of the gapless SO(2) fluctuations of $\mathbf{\Omega}_{\mathrm{f}}$ in the superconducting state.

The SO(2) fluctuations of $\mathbf{\Omega}_{\mathrm{f}}$ are well described by a non-linear sigma model if the energy gap to the electronic (amplitude) excitations is sufficiently large. Near the phase transition the coupling to the amplitude excitations plays an important role.[17] The crossover between the two regions defines the crossover gap $\Delta_c$.

Only for $|\mathbf{\Delta}_T(H)| > \Delta_{\mathrm{c}}$ the gapless flavor fluctuations[42] of the order parameter components can be described by the 2+1 dimensional non-linear sigma model as discussed in detail in Ref. 17. For $|\mathbf{\Delta}_T(H)| < \Delta_{\mathrm{c}}$ the system is in the "effective saddle point" regime where the amplitude mode energy cutoff plays a crucial role and the internal degrees of freedom are integrated out. The field at which the order parameter is equal to the crossover gap can be identified as $H_{\mathrm{p}}$, i.e., $|\mathbf{\Delta}_T(H_{\mathrm{p}})| = \Delta_{\mathrm{c}}$. For smaller fields $H < H_{\mathrm{p}}$ one has $|\mathbf{\Delta}_T(H)| > \Delta_{\mathrm{c}}$ and the system exhibits gapless flavor fluctuations which increase[23,24] the magnetic response through the spin-flavor coupling.

In order to derive the specific geometry and temperature dependence of $H_{\mathrm{p}}$ within the framework of the present approach consider that from Eqs. (14) – (16) and $H_{\mathrm{c},u} \gg H_{\mathrm{c},a}$ follows that the modulus of the order parameter is

$$\frac{|\mathbf{\Delta}_T(H,\phi)|^2}{\Delta_0^2} \approx \frac{\left[H_{\mathrm{c},u}(T) - H\right]^2 + H_{\mathrm{c},a}^2(T) f_1^2(\sin 2\phi)}{H_{\mathrm{c},u}^2(0)} \,. \tag{19}$$

The crossover field $H_{\mathrm{p}}(\phi)$ is defined by $|\mathbf{\Delta}_T(H_{\mathrm{p}})| = \Delta_c$ and takes a simple form in the case $\phi = 0$ or, equivalently, for $H_{\mathrm{c},a}(T) = 0$:

$$H_{\mathrm{p}}(0) = H_{\mathrm{c},u}(T) - \frac{H_{\mathrm{c},u}(0)}{\Delta_0}\ \Delta_{\mathrm{c}} \,. \tag{20}$$

The crossover field $H_{\mathrm{p}}$ is shifted by a temperature independent constant with respect to $H_{\mathrm{c},u}(T)$.

For $H_{\mathrm{c},a}(T) \neq 0$ and $\phi \neq 0$ the SO(2) fluctuations between the order parameter components $\Delta_{T,x}$ and $\Delta_{T,y}$ only are possible if the ratio

$$r_\Delta^2 = \frac{|\Delta_{T,x}^2 - \Delta_{T,y}^2|}{|\mathbf{\Delta}_T|^2} \tag{21}$$

is small enough, i.e., $r_\Delta^2 < r_{\mathrm{c}}^2$. The theoretical form of the angular dependent field then becomes

$$H_{\mathrm{p}}(\phi) = H_{\mathrm{c},u}(T) - \frac{2H_{\mathrm{c},a}(T)}{r_c^2}\ |f_1(\sin 2\phi)| - \sqrt{\frac{H_{\mathrm{c},u}^2(0)}{\Delta_0^2}\ \Delta_{\mathrm{c}}^2 - H_{\mathrm{c},a}^2(T) f_1^2(\sin 2\phi)} \,. \tag{22}$$

The resulting fit to the experimental data at $T = 0.06$ K is shown as the dotted line in Fig. 3 and yields the parameters $\Delta_{\mathrm{c}}(T \to 0) = 0.4$ K and $r_{\mathrm{c}}^2 = 0.64$. The agreement with the experimental results is quite satisfactory.

### C. Temperature dependence

Figure 4(a) and (b) show the temperature dependence of the fields $H_{\mathrm{c2}}$, $H_2$ and, $H_{\mathrm{p}}$ as derived from ac-susceptibility measurements (Ref. 24) for $\phi = 45°$ and $\phi = 0°$, respectively. The temperature dependence of $H_{\mathrm{c},u}(T) = [H_{\mathrm{c2}}(T) + H_2(T)]/2$ is essentially given by Eq. (3). From the difference $H_{\mathrm{c2}} - H_2 = 2H_{\mathrm{c},a}$ at $\phi = 45°$ the temperature dependence of the anisotropic field component can be extracted as

$$H_{\mathrm{c},a}(T)\big|_{T \leq T_{\mathrm{c},a}} \approx H_{\mathrm{c},a}(0)\ \left(1 - \frac{T^2}{T_{\mathrm{c},a}^2}\right) \tag{23}$$

with $H_{\mathrm{c},a}(0) = 0.047$ T, $T_{\mathrm{c},a} = 0.85$ K and $H_{\mathrm{c},a}(T)|_{T > T_{\mathrm{c},a}} = 0$. The experimental data (symbols) and the fit (full line) are shown in Fig. 4(c). From Eqs. (14) and (15) follows that the difference $\Delta_{T,x} - \Delta_{T,y} \approx 0.2$ K for $\phi = 45°$ ($H \leq H_2$). Consequently the vanishing of $H_{\mathrm{c},a}(T)$ for $T > T_{\mathrm{c},a}$ can be associated with thermal fluctuations obscuring the difference between the order parameter components $\Delta_{T,x}$ and $\Delta_{T,y}$.

The symbols in Fig. 4(d) show the data for $H_{\mathrm{c},u}(T) - H_{\mathrm{p}}(\phi, T)$ as extracted from the experimental data for $\phi = 45°$ (circles) and $\phi = 0°$ (squares). The broken line is the temperature independent result from Eq. (22) for $\phi = 0°$ with $\Delta_{\mathrm{c}} = 0.3$ K in excellent agreement with experiment. The discrepancy between the values extracted here [$\Delta_{\mathrm{c}} = 0.3$ K and $H_{\mathrm{c},a}(0) = 0.047$ T] and the values extracted from the angular dependence shown in Fig. 3 [$\Delta_{\mathrm{c}} = 0.4$ K and $H_{\mathrm{c},a}(0) = 0.05$ T] can in part be attributed to experimental uncertainties and in part to the corrections to the theory for $T < 0.4$ K as anticipated in Sec. II B.

For $\phi = 45°$ the experimental data [circles in Fig. 4(d)] are only in qualitative agreement with the fit form Eq.

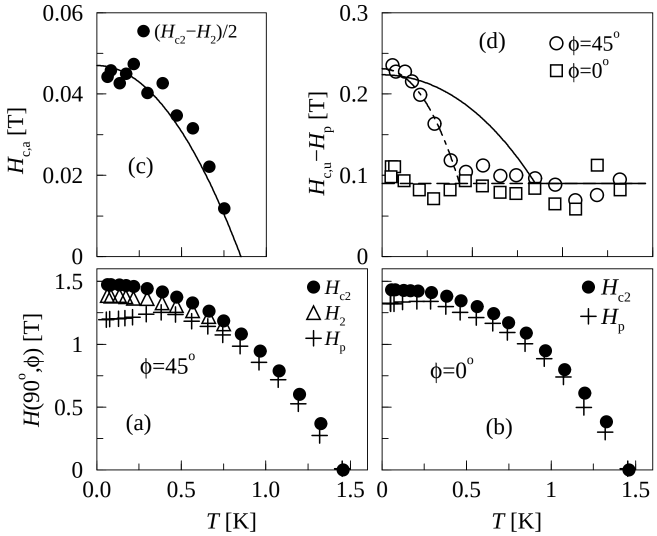


FIG. 4. *Panels (a) and (b) show the temperature dependence of the critical in-plane fields as derived from ac-susceptibility measurements*[24] *for* $\phi = 45^\circ$ *and* $\phi = 0^\circ$*, respectively. Panel (c) shows* $H_{c,a} = (H_{c2} - H_2)/2$ *together with a fit from Eq. (23). Panel (d) shows* $H_{c,u}(T) - H_p(\phi, T)$ *from experimental data together with the respective fits from Eqs. (22) and (23) for* $\phi = 45^\circ$ *(circles, full line) and* $\phi = 0^\circ$ *(squares, dashed line) with* $H_{c,a}(0) = 0.047$ *T and* $T_{c,a} = 0.85$ *K. The dash-dotted line is obtained using* $H_{c,a}(0) = 0.05$ *T and* $T_{c,a} = 0.43$ *K for* $\phi = 45^\circ$*.*

(22) [full line, $\Delta_c = 0.3$ K, $r_c^2 = 0.64$, $H_{c,a}(0) = 0.047$ T, $T_{c,a} = 0.85$ K] for 0.2 K $< T <$ 0.8 K. This might indicate that there are corrections to the theory. On the other hand the read-off from the ac-susceptibility data of $H_p$ for $T <$ 0.6 K and of $H_2$ for $T >$ 0.6 K is not unambiguous.[24] To match the experimental data in Fig. 4(d) parameters $H_{c,a}(0) = 0.05$ T and $T_{c,a} = 0.43$ K are adequate [dash-dotted line in Fig. 4(d)]. A more complex temperature dependence with a small $H_{c,a}(T) < 0$ for $T > 0.7T_c$ has been proposed in Refs. 20,23. An angular analysis of the ac-susceptibility as shown in Fig. 3 at temperatures 0.2 K $< T <$ 0.8 K would be helpful. Complimentary data from specific heat measurements as a function of the in-plane field direction are also desirable.

The crossover or "fluctuation field" $H_p$ marks the appearance of quasi two-dimensional fluctuations as described by the non-linear sigma model. In that sense $H_p$ marks a phenomenon similar to a Kosterlitz-Thouless transition.[43]

### D. Interpretation of $\Delta_c$ in terms of the microscopic model

For reduced temperatures $t \le t_c \approx 0.05$ the order parameter fluctuations are in the "effective saddle point" regime as defined in Ref. 17. In the effective saddle point regime the superconducting gap defines a cutoff in the spectrum of the gapless SO(2) fluctuations of the internal flavor degrees of freedom of the order parameter.[42] Gapless fluctuations of the order parameter components as described by the 2+1 dimensional non-linear sigma models without an energy cutoff only exist for $t(H \to 0) > t_c$.

The analogy between the reduced field $h$ and the reduced temperature $t$ (Sec. II B) implies that for $h(T \to 0) \le h_c \approx 0.05$ the system also is in the effective saddle point regime. The value of $\Delta_c \approx 0.4$ K (Sec. III B) gives together with Eq. (12) $h_c(T \to 0) \approx 0.08$. Considering the experimental uncertainties and the simplicity of the approach this agreement is quite satisfactory. Equation (12) can be rewritten as

$$h_c(T) = \frac{\Delta_c}{\Delta_0} \, \frac{H_{cm}(0)}{H_{cm}(T)} \tag{24}$$

and reveals the temperature dependence of $h_c(T)$.

Similarly follows from Eq. (11) that with $\Delta_c \approx 0.3$ K (Sec. III C) $t_c(H \to 0) \approx 0.06$. Again, the agreement with the proposed value of $t_c \approx 0.05$ underlines the consistency of the microscopic model in Refs. 16,17 and the phenomenological approach herein.

The analysis discussed herein suggests that the maximum in the specific heat below the superconducting transition coincides with $H_p$. Probing the specific heat dependent on the in-plane field direction experimentally would be an appropriate test of this prediction.

### E. Out-of-plane spin-flip field $H_\perp$ and out-of-plane anisotropy of $H_{c,a}$

At low temperatures and for small out-of-plane fields $H(0,\phi) < H_\perp \| H(0,\phi)$ the thermodynamic and transport properties of the system are basically unaffected. The value $H_\perp \approx 0.01$ T can be determined both from the specific heat[14] as well as from thermal conductivity[39,37] measurements. Obviously there is a small confinement of the magnetic moment of the Cooper pairs to the $x$-$y$ plane that becomes apparent when fluctuations are frozen out. The spin-flip field $H_\perp$ is found to be temperature independent as long as $H_{c2}(0,\phi,T) > H_\perp$[39,37] and shows a temperature dependent hysteresis.[37]

The dependence of the field $H_2(\theta, 45^\circ)$ on the polar angle $\theta$ as observed in Ref. 24 is closely related to the out-of-plane spin-flip field. For $\theta < 89.5^\circ$ the out-of-plane component of the magnetic field $\hat{\boldsymbol{z}}\boldsymbol{H}$ is larger than the spin-flip field, i.e., $\hat{\boldsymbol{z}}\boldsymbol{H} > H_\perp$. For $\theta < 89.5^\circ$ the order parameter has an out-of-plane spin component which couples homogeneously to both spatial components $\Delta_{T,x}(H)$ and $\Delta_{T,y}(H)$. $H_{c,a}$ vanishes and $H_2 = H_{c2}$. The transition from $H_{c,a}(\theta > 89.5^\circ) \neq 0$ to $H_{c,a}(\theta < 89.5^\circ) = 0$ should be of first order for $T \le 0.02$ K.

## IV. SUMMARY

Based on the microscopic model introduced in Refs. 16 and 17 the observed specific heat and ac-susceptibility data in the superconducting phase in $Sr_2RuO_4$ with applied magnetic fields have been described consistently.

### A. Conclusions

(i) The temperature dependence of the upper critical field is satisfactorily described by a phenomenological formula similar to the BCS results [Eq. (3)].

(ii) The dependence of the upper critical field on the out-of-plane angle of the field direction is excellently reproduced by the Landau-Ginzburg anisotropic effective mass approximation if the enhanced in-plane coupling is included [Eq. (5)].

(iii) The specific heat below the superconducting phase transition increases linearly with decreasing temperature as predicted in the presence of quasi two-dimensional gapless fluctuations of the order parameter. The observed reduction of the temperature slope with increasing magnetic field is consistent with the reduced magnetic degrees of freedom. The resulting linear dependence of the specific heat on the magnetic field just below the critical field is temperature independent and in quantitative agreement with experiment.

(iv) The two spatially anisotropic components of the order parameters couple differently to the applied magnetic field depending on its orientation. The observed angular dependence of the resulting two critical fields leads to conclude that each of the components of the order parameter has a spatial anisotropy of $\sim 7$ % for $H \to 0$.

(v) The non-analytic angular variation of the in-plane upper critical fields supports the presence of quasi one-dimensional correlations along the diagonals of the basal plane of the unit cell as predicted by the underlying microscopic model.

(vi) The order parameter fluctuations are described for fields $H \ll H_{\mathrm{p}}$ by the non-linear sigma model while for $H_{\mathrm{p}} < H < H_{\mathrm{c2}}$ they are qualitatively altered by the coupling to the amplitude fluctuations. Both the angular and temperature dependence of the fluctuation field $H_{\mathrm{p}}$ are described consistently with ac-susceptibility measurements.

(vii) For small temperatures $T < 0.1$ K the spin-gap field $H_{\mathrm{sg}} \approx 0.12$ T has been determined. For fields $H < H_{\mathrm{sg}}$ the spectrum of the internal magnetic degrees of freedom of the order parameter $\overline{\mathbf{\Omega}}_{\mathrm{s}}$ has a gap mediated by spin-flavor coupling. For $H = 0$ the gap (Leggett frequency) has been estimated as $\Omega_A \sim 0.08$ K.

### B. Proposed experiments

For further clarification of the description of the low temperature electronic properties of $Sr_2RuO_4$ the following additional experiments would be useful.

(i) An angular analysis of the ac-susceptibility at temperatures $0.2\,\mathrm{K} < T < 0.8\,\mathrm{K}$ would be helpful to clarify if there are corrections to the theory concerning the angular dependence of the field $H_{\mathrm{p}}$ in that temperature range.

(ii) Complimentary data from specific heat measurements as a function of the in-plane field direction are desirable to test if the maximum of the specific heat coincides with $H_{\mathrm{p}}$.

(iii) Specific heat measurements in the range of $0 < T < 0.4$ K and $1.4 < H < 1.5$ T are desirable to study details of the slope of the linear field and temperature dependence of the specific heat near the transition. Possible corrections to the theory in that parameter range can then be investigated.

(iv) Excess current measurements provide a quantitative test of the linear dependence of the order parameter on the temperature and the magnetic field as predicted in (11) and (12).

### C. Critique and outlook

The aim of the presented analysis of the properties of $Sr_2RuO_4$ in magnetic fields is to consistently describe a large number of experimental results within a minimal model. To this end two assumptions where made. The first is the separation of the magnetic and flavor degrees of freedom as imposed by the underlying microscopic model discussed in Ref. 16 and 17. Secondly it is assumed that the magnetic field dependence of the order parameter can be described by simple relations similar to the temperature dependence as given in Eqs. (11) and (12). The qualitative and quantitative agreement of the specific heat data (Secs. II B and II C) and the geometry of the critical fields (Secs. III A, III B and III C) underline the validity of the approach.

Based on these results a more detailed study of the interplay of the magnetic and flavor degrees of freedom together with the vortex lattice via a full Landau Ginzburg analysis appears desirable. Similar approaches were performed earlier for a two-component $p$-wave order parameter[20,21,44] and $^3$He.[45,32]

## ACKNOWLEDGMENTS

I thank M. Eschrig, J. Kroha, and F. Laube for discussions. The work was supported by DOE contract number DE-AC02-98CH10886 and the Center for Functional Nanostructures at the University of Karlsruhe.

[1] Y. Maeno *et al.*, Nature (London) **372**, 532 (1994).
[2] Y. Maeno, T. M. Rice, and M. Sigrist, Physics Today **54**, 42 (2001).
[3] T. M. Rice and M. Sigrist, J. Phys.: Condens. Matter **7**, L643 (1995).
[4] M. Braden *et al.*, Phys. Rev. B **57**, 1236 (1998).
[5] T. Oguchi, Phys. Rev. B **51**, 1385 (1995).
[6] A. P. Mackenzie *et al.*, Phys. Rev. Lett. **76**, 3786 (1996).
[7] M. Schmidt *et al.*, Phys. Rev. B **53**, R14761 (1996).
[8] Y. Maeno *et al.*, J. Phys. Soc. Jpn. **66**, 1405 (1997).
[9] A. P. Mackenzie *et al.*, J. Phys. Soc. Jpn. **67**, 385 (1998).
[10] K. Ishida *et al.*, Phys. Rev. Lett. **84**, 5387 (2000).
[11] A. V. Puchkov, Z.-X. Shen, T. Kimura, and Y. Tokura, Phys. Rev. B **58**, R13322 (1998).
[12] S. Nishizaki, Y. Maeno, and S. Farner, J. Phys. Soc. Jpn. **67**, 560 (1998).
[13] S. Nishizaki, Y. Maeno, and Z. Q. Mao, J. Low Temp. Phys. **117**, 1581 (1999).
[14] S. Nishizaki, Y. Maeno, and Z. Q. Mao, J. Phys. Soc. Jpn. **69**, 572 (2000).
[15] M. Suzuki *et al.*, Phys. Rev. Lett. **88**, 227004 (2002).
[16] R. Werner and V. J. Emery, cond-mat/0208306 (2002).
[17] R. Werner, cond-mat/0208307 (2002).
[18] D. F. Agterberg, V. Barzykin, and L. P. Gor'kov, Phys. Rev. B **60**, 14868 (1999).
[19] G. Baskaran, Physica B **223-224**, 490 (1996).
[20] M. Sigrist, J. Phys. Soc. Jpn. **69**, 1290 (2000).
[21] D. F. Agterberg, Phys. Rev. B **64**, 052502 (2001).
[22] K. Yoshida, Y. Maeno, S. Nishizaki, and T. Fujita, Physica C **80**, 161 (1996).
[23] Z. Q. Mao *et al.*, Phys. Rev. Lett. **84**, 991 (2000).
[24] H. Yaguchi *et al.*, submitted to Phys. Rev. B (2002), cond-mat/0106491.
[25] G. M. Luke *et al.*, Nature (London) **394**, 558 (1998).
[26] F. Laube *et al.*, Phys. Rev. Lett. **84**, 1595 (2000).
[27] P. Ehrenfest, Comm. Phys. Lab. Univ. Leiden Suppl. **75b**, I,1 (1933).
[28] T. Akima, S. Nishizaki, and Y. Maeno, J. Phys. Soc. Jpn. **68**, 694 (1999).
[29] Z. Mao, Y. Mori, and Y. Maeno, Phys. Rev. B **60**, 610 (1999).
[30] N. R. Werthamer, E. Helfand, and P. C. Hohenberg, Phys. Rev. **147**, 295 (1966).
[31] M. Tinkham, *Introduction to superconductivity*, *2nd edition* (McGraw-Hill, New York, 1996).
[32] G. E. Volovik, *Exotic properties of superfluid* $^3$*He*, *Series in Condensed Matter Physics, Vol.1* (World Scientific, Singapore, 1992).
[33] In $^3$He-$A$ the presence of spin-orbit coupling yields a gap in the magnetic spectrum given by the Leggett frequency[45,32] $\Omega_A^2 \approx a^2\overline{v}_{\rm eff}^2/\xi_{\rm P}^2$. The analogy here is the spin-flavor coupling. An estimate is obtained via $\xi_{\rm P}/a \sim 250$ [K. Yoshida, Y. Maeno, S. Nishizaki, and T. Fujita, Physica C **80**, 161 (1996); A.P. Mackenzie *et al.*, Phys. Rev. Lett. **80**, 161 (1998)] and $\overline{v}_{\rm eff} \sim 22$ K as $\Omega_A < 0.1$ K.
[34] P. G. Kealey *et al.*, Phys. Rev. Lett. **84**, 6094 (2000).
[35] R. Heeb and D. F. Agterberg, Phys. Rev. B **59**, 7076 (1999).
[36] M. A. Tanatar *et al.*, Phys. Rev. B **63**, 064505 (2001).
[37] M. Suzuki *et al.*, J. Phys.: Condens. Matter **14**, 7371 (2002).
[38] M. A. Tanatar *et al.*, Phys. Rev. Lett. **86**, 2649 (2001).
[39] K. Izawa *et al.*, Phys. Rev. Lett. **86**, 2653 (2001).
[40] R. Werner, cond-mat/0208301 (2002).
[41] A. Klümper, Euro. Phys. J. B **5**, 677 (1998).
[42] The fluctuations of the internal magnetic degrees of freedom of the order parameter are assumed to be frozen out by the magnetic field, c.f. Sec. II C.
[43] A. M. Tsvelik, *Quantum field theory in condensed matter physics* (Cambridge University Press, Cambridge, 1995).
[44] E. Babaev, Phys. Rev. Lett. **88**, 177002 (2002).
[45] G. E. Volovik and M. V. Khazan, Sov. Phys. JETP **58**, 551 (1983).